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ORIGINAL PAPER

Assessment of Cu and Zn contamination and associated human health risks in urban soils from public green spaces in the city of Thessaloniki, Northern Greece

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Abstract

Sustainable patterns of production and consumption dictate waste elimination and the modest use of resources in uncontaminated environments. However, this is not always possible, especially in urban settings, since soils may be enriched with various metals and metalloids, and this may pose a threat to the city's residents. Urban soils have not received the same degree of interest as agricultural soil, where crops are cultivated, but the risk of exposure of humans to soil pollutants is real in urban settings. For this reason, in the present study, soil samples from public green spaces in Thessaloniki, Northern Greece, were collected in March 2022. Soil physicochemical characteristics (% of sand, silt and clay, pH and electrical conductivity) as well as the pseudo-total concentrations of Cu and of Zn (mg/kg dry weight soil) were determined for 50 distinct sites in the city. The risk to inhabitants was determined for the oral and dermal routes in both adults and children. The pseudo-total concentrations of Cu and of Zn were variable, with some sites exhibiting concentrations higher than the EU set limits for soil treated with municipal sludge or than the Canadian Soil Quality Guidelines. The risk to adults and children was acceptable in every case. The high correlation between the Cu and Zn pseudo-total concentrations as well as the locations of the most polluted sites point to input from anthropogenic activities. These activities may include train and automobile traffic, as well as operations within the city's commercial and passenger port. Even though it is not as well studied as soil pollution in agriculture, soil pollution in cities should be given proper attention.

Keywords Urban soil pollution · Risk assessment · Cu · Zn · Pollution sources

Introduction

Modern societies strive for continuous economic development which should diffuse to all social strata and respect the environment. Sustainable methods of production and consumption offer viable solutions to the simultaneous environmental crises of climate change, biodiversity loss and raw resource diminishment. Nowadays, cyclic patterns of production and consumption dictate waste elimination and the reuse of any disposed material that can be reshaped; for example, the use of organic waste as fertilizer to enhance crop production (Almendro-Candel et al. 2018; Rodríguez-Espinosa et al. 2023). Soil degradation and soil pollution may, however, jeopardize this sustainable

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production. Specifically, the presence of heavy metals or potentially toxic elements (PTEs) in the agro-environment has been the subject of numerous studies in recent decades due to their serious effects on human health (Alloway 2013; Singh et al. 2018). Unlike other toxic elements, heavy metals and metalloids are not degraded by natural soil processes, and they may bioaccumulate in organisms (Kabata-Pendias 2011). Thus, heavy metal pollution poses a significant threat to human health and well-being (Bourliva et al. 2021; Aslanidis and Golia 2022). This has been verified for soils that are used for agricultural purposes (Ben Chabchoubi et al. 2020; Navarro-Pedreño et al. 2018; Smaoui et al. 2020); however, many other types of soil are also susceptible to this kind of pollution (Stylianou et al. 2020). Numerous mechanisms determine the mobility and therefore the availability of metals in the soil, while adsorption to the solid soil surface depends on the physicochemical properties of the soil along with the environmental conditions of the study area (Dimirkou et al. 2009; Golia et al. 2023).

Urban soil pollution has received considerably less attention; however, good soil quality is vital for human health and well-being (Rodríguez-Espinosa et al. 2021). Furthermore, when compared to adjacent undisturbed soils, urban soils appear to be quite contaminated (Ajmone-Marsan and Biasioli 2010). PTEs in urban soils derive from both natural-geochemical and anthropogenic sources (Golia et al. 2021). The production, use and distribution of metallic and electronic products or chemical and pharmaceutical products as well as the use of fertilizers in urban agriculture are only some of the human activities that contaminate urban soils (Antoniadis et al. 2019). Furthermore, the use of motor vehicles and their combustion of fossil fuels produce fragments that end up in topsoil, e.g., worn tires and brake pads as well as elements found in the combustible material (Peng et al. 2022). Mediterranean urban soils can be the sink for the increased air pollution from Northwestern Europe all year long and the recipient of tourism-related pollution insurgence in the summer months (Papamichael et al. 2022; Zafeirakou et al. 2022). These threats are in stark contrast to the aspiration for "environmentally data-driven smart sustainable cities" of the future (Bibri 2020) where environmental sustainability is ensured.

The present manuscript assesses the anthropogenic enrichment of urban soils in the conglomeration of Thessaloniki, the most populated city in Northern Greece. The sites examined correspond to public open spaces where multiple land uses are present (Guilland et al. 2018) and they belong to public parks, playgrounds and sports fields, cemeteries, and archeological sites, or they act as road dividers or railway line substrates. It should be noted that soil in urban areas is mostly entrenched by infrastructure such as buildings, roads and pavements, and thus it cannot be studied as a continuous system. The PTEs examined here were Cu and Zn, which are some of the least toxic metals that may be found in soil due to anthropogenic input. Cu may build up in the human body, targeting mostly the gastrointestinal and hepatic systems at high concentrations (ATSDR 2022). Large doses of Zn may cause stomach cramps, nausea, and vomiting, while subchronic exposure may cause anemia, damage the pancreas, and decrease levels of high-density lipoprotein cholesterol (ATSDR 2005). These metals were chosen as part of a preliminary study to establish the background contamination which may be ubiquitous in the city. As such, an initial snapshot of the current contamination of Thessaloniki urban soils has been created, revealing polluted "hotspots" that may need further consideration.

Materials and methods

Description of the sampling area

Thessaloniki (40° 62′ E, 22° 95′ N) is the second largest city in Greece and a commercial, transport and cultural center in the Balkans. With a population of 317,778 permanent residents (ELSTAT 2021), the city center is characterized by a high building factor and high road traffic, especially in the morning and early evening hours, with over 400,000 vehicles on major and minor transportation routes. Thessaloniki's port, located in the west of the city, offers both passenger and commercial cargo transportation (Bourliva et al. 2021). The New Thessaloniki Railway Station is located in the central quarter of Xirokrini on Monastiriou Street and provides transportation to various cities through the country by electric trains. Sampling was performed in public green spaces scattered across the city center. Figure 1 shows the sampling points (X1–X50). The points closest to the railway station are shown in the upper left corner.

The sampling points were in areas accessed and utilized by several people per day, such as the AUTH campus, hospital surroundings, and public spaces such as playgrounds, schoolyards and recreational parks. Each green area used for this study was between 25 and 100 m^2 in size. Sampling was performed during dry periods in March 2022.

Chemical analysis of soil samples

Surface soil sampling (0–20 cm) was carried out with a wooden shovel. At least three samples were taken from each area, and each sample consisted of four sub-samples within a radius of approximately 1 m from the sampling point. The soil samples were placed in plastic bags and transported to the AUTH soil science laboratory. There, they were air dried at room temperature and then sieved using a 2-mm-diameter pore sieve. Three replicate subsamples of 600 g were taken from each sample for further physicochemical analyses,

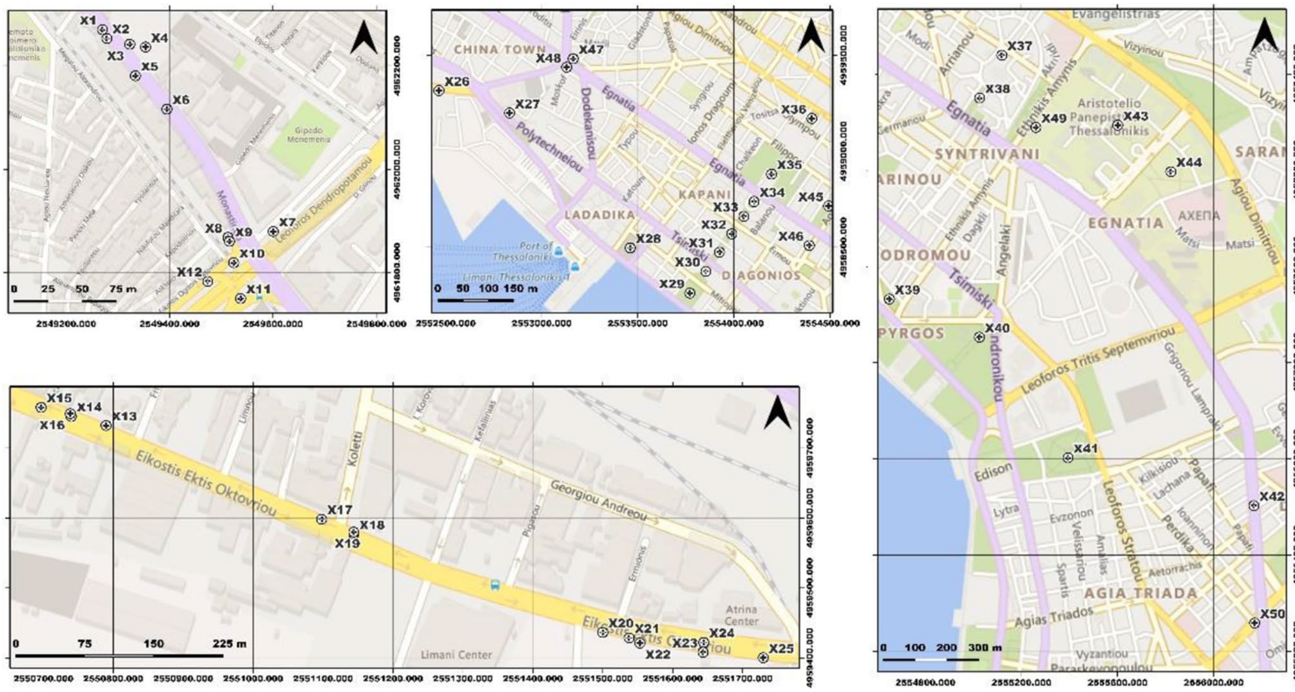


Fig. 1 Sampling points in Thessaloniki City

as described by Page et al. (1982). The texture of the soil was determined by the Bouyoucos method as described in Berreta et al. (2014) after determining the percentage (%) of sand, silt and clay. The pH value was determined in a soil suspension with a soil water ratio of 1:2 (w/v). In the same soil suspension, the electrical conductivity value was also determined, and the results were expressed in μS cm^{-1}. To determine the concentrations of metals, 1 g of soil and 12 mL aqua regia (9 mL HCl + 3 mL HNO_3) were added to the appropriate autoclave device with a Teflon bomb (Labter, Ecopre Series, AHF, Germany), and extraction was carried out using the aqua regia method (ISO 1994) The system followed a digestion program in which the temperature gradually reached 180 °C and the total digestion time was 4.5 h. Then, in the extracts, the pseudo-total concentrations of Cu and Zn were determined using an atomic absorption spectrophotometer (Shimadzu, AA-6600, Duisburg, Germany) with flame and graphite furnace equipment at 325 and 213.9 nm, respectively (Williams 1984). A certified soil sample (BCR-146R) was used, and the percentage recovery was 91.9% and 95.3% for Cu and Zn, respectively.

Calculation of human risk due to exposure to Cu and Zn

The recommendations of the USEPA for human non-carcinogenic risk assessment, as laid out in *Risk Assessment Guidance for Superfund* (USEPA 1989), were followed. The resident scenario (adults and children) was chosen since the areas are within the urban agglomeration. Both ingestion and dermal routes were deemed relevant, while the inhalation risk, such as that measured in Papaoikonomou et al. (2018), cannot be readily calculated since the inhalable fraction is unknown. The hazard quotient (HQ) was calculated as follows (Singh and Kumar 2017):

$$HQ_{ingestion} = \frac{CDI_{ingestion}}{RfD_{ingestion}} \tag{1}$$

$$HQ_{dermal} = \frac{CDI_{dermal}}{RfD_{dermal}}, \tag{2}$$

where $RfD_{ingestion}$ and RfD_{dermal} are the reference doses for each metal via the oral and dermal routes, respectively, and $CDI_{ingestion}$ and CDI_{dermal} are the chronic daily intake of each metal via the oral and dermal routes, respectively. The $RfD_{ingestion}$ values for Cu and for Zn were taken from Peña-Fernández et al. (2014), while the RfD_{dermal} for Cu was taken from RAIS (2022), based on a gastrointestinal absorption factor of 0.3. Zn is not considered toxic through the dermal route.

The parameters $CDI_{ingestion}$ and CDI_{dermal} were calculated according to USEPA (1989) as amended by Antoniadis et al (2019) and to USEPA (1989) as amended by Singh and Kumar (2017), respectively, as follows:

$$\mathrm{CDI}_{\mathrm{ingestion}} = \frac{\mathrm{CS} \times \mathrm{IR} \times \mathrm{CF} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}}, \quad (3)$$

where CS = chemical concentration in soil (mg element/kg dry soil, given in Fig. 1), IR = ingestion rate (100 mg/d for adults, 200 mg/d for children), CF = conversion factor (10^{-6} kg/mg), EF = exposure frequency (365 d), ED = exposure duration (70 y for adults, 6 y for children), BW = body weight (70 kg for adults, 15 kg for children), AT = averaging time (70 y × 365 d/y for adults, 6 y × 365 d/y for children);

$$\mathrm{CDI}_{\mathrm{dermal}} = \frac{\mathrm{CS} \times \mathrm{CF} \times \mathrm{SA} \times \mathrm{AF} \times \mathrm{ABS} \times \mathrm{ET} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}}, \quad (4)$$

where CS = chemical concentration in soil (mg element/kg dry soil, given in Fig. 1), CF = conversion factor (10^{-6} kg/mg), SA = skin surface area (the average for adult males and females is 18,200 cm^2, the average for male and female children is 7200 cm^2), AF = soil to skin adherence factor (1.45 mg/cm^2), ABS = chemical specific value (0.03 from Singh and Kumar 2017), EF = exposure frequency (365 d), ET = exposure time (0.6 from Singh and Kumar 2017), ED = exposure duration (70 y for adults, 6 y for children), BW = body weight (70 kg for adults, 15 kg for children), AT = averaging time (70 y × 365 d/y for adults, 6 y × 365 d/y for children).

The total non-carcinogenic risk for each site in each scenario (oral or dermal, adults or children) was calculated according to the USEPA as the sum of the individual HQs:

$$\mathrm{HI} = \Sigma \mathrm{HQ}_i, \quad (5)$$

where HI = hazard index and i indicates the metal considered.

Statistics

Graphs and figures were plotted in Microsoft Excel 365 (Microsoft Corp, Washington, USA). Possible correlations between the parameters pH, EC, % silt, sand or clay, and concentration of Cu or Zn in soil were examined through the Spearman correlation in SPSS28 (IBM, Armonk, USA).

Results and discussion

Some physicochemical characteristics of the soil samples as well as the Cu and Zn concentrations are shown in Table 1. As shown, there are variations in soil pH, with values ranging from neutral to more basic. Mean electrical conductivity (EC) was approximately 245 µS/cm, although an extreme value of 1431.50 µS/cm was also found. Soil composition varied from sandy samples (71.20%) to heavy clay soils (36.60%). Even though it was not measured, the soil was visually found to have a poor organic matter content, as expected for compacted urban soils (Navarro-Pedreño et al. 2021).

Finally, the pseudo-total concentrations of Cu and of Zn varied greatly, with some samples exhibiting minimal presence of these metals. However, there were some sampling points with high concentrations that could not be attributed to a natural background. This is clearly shown in Fig. 2, where one sample for Cu (Fig. 2A) and two samples for Zn (Fig. 2B) were above the EU set limits (140 mg/kg d.w.

Table 1 General physicochemical characteristics of the soils and pseudo-total metal concentrations (mg/kg dw soil) compared to the standard limits for Cu and Zn (values are the mean ± standard deviation from three replicates)

	pH	EC (µS/cm)	Clay (%)	Sand (%)	Silt (%)	Zn (mg/kg)	Cu (mg/kg)
Descriptive statistics							
Minimum value	7.75 ± 0.05	77.80 ± 1.33	10.80	36.80	10.20	21.34 ± 0.42	17.00 ± 1.93
Maximum value	9.17 ± 0.02	1431.50 ± 11.5	36.60	71.20	36.00	402.00 ± 2.81	138.30 ± 9.57
Mean value	8.42	244.66	20.73	54.83	24.42	95.11	86.87
Standard deviation	0.39	203.03	6.68	9.36	5.20	68.38	83.14
Recommended upper limits							
Directive 86/278/EEC						300	140
Canadian SQGs[a]						250	63
Brazilian SQGs[b]						1000	400
Other SQGs[c]						140[d]	120[e]

[a]From CCME (2022)

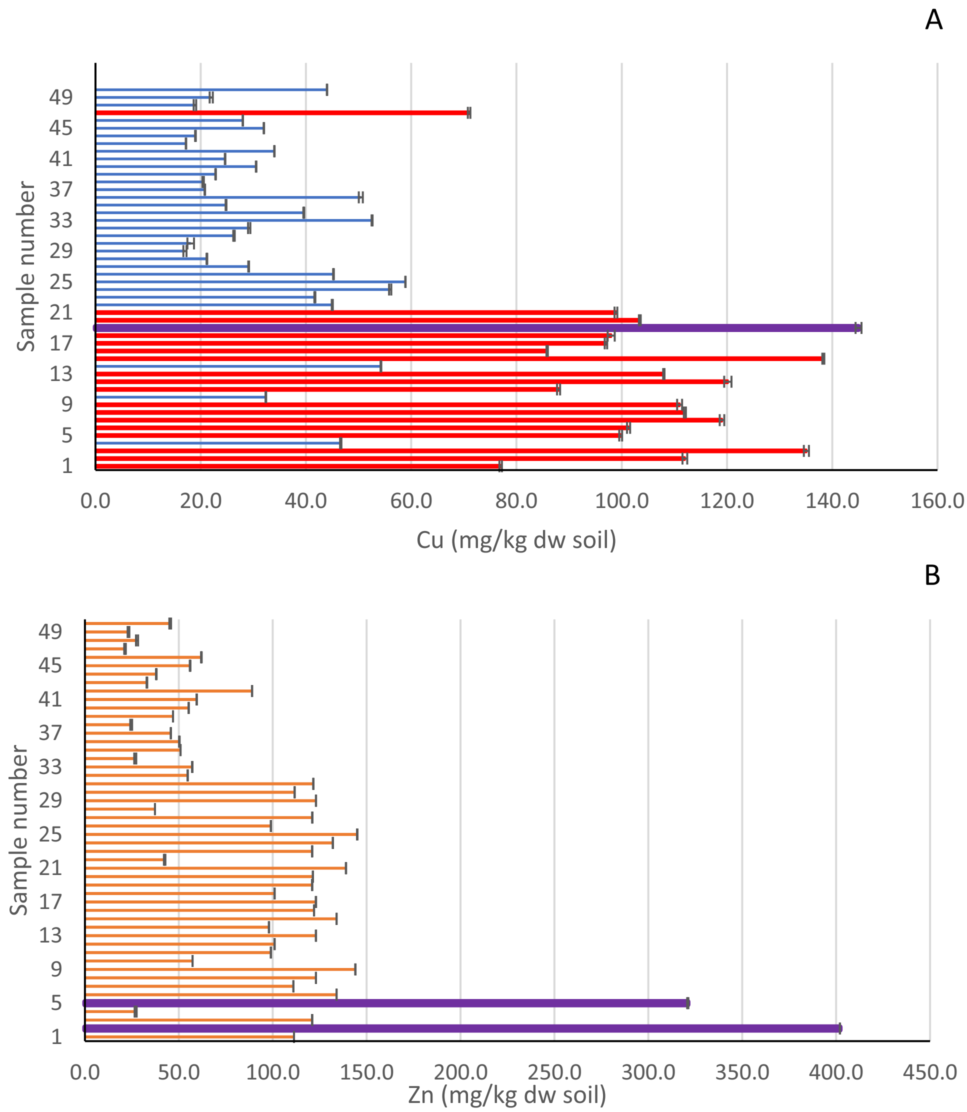
[b]From Lima et al. (2023)

[c]From Ajmone-Marsan and Biasioli (2010)

[d]According to Dutch legislation

[e]According to Italian legislation

Fig. 2 Pseudo-total concentrations ofCu (**A**) and Zn (**B**) in samples ($n=50$). Values above the EU set limits are shown as *thick purple lines* and values above the Canadian SQGs are shown as *red lines*. Values are the mean ± standard error of the mean ($n=3$ replicates)



soil and 300 mg/kg d.w. soil, respectively) for soil treated with municipal sewage sludge (CEC 1986). Less universal limits apply in the case of urban soil; as such, our results were compared with relevant guidelines on human health. The Canadian SQGs (Soil Quality Guidelines) comprise specific limits for the protection of environmental and human health in residential and parkland areas (CCME 2022). For the present campaign, 18 samples were above the set limit for Cu (Fig. 2A) and 2 samples were above the set limit for Zn (Fig. 2B) in the Canadian SQGs. According to the Brazilian Ministry of Environment, the limit values for residential exposure to Cu and Zn in soil are 400 mg/kg d.w. and 1000 mg/kg d.w., respectively (Lima et al. 2023). Under these lenient guidelines, no samples from the present campaign were above the limit. For two EU countries (Italy and the Netherlands), the limit "for remediation of residential areas" is 120 mg Cu/kg d.w. and the limit "for sustainable soil quality" is 140 mg Zn/kg d.w., respectively (Ajmone-Marsan and Biasioli 2010). For these comparisons, 14 and 4 samples were above the set limits for Cu and Zn, respectively.

Even though it is less informative, the mean values of Cu and of Zn found here can be compared to the values of these metals found in other urban settings. As such, Cu at 86.9 mg/kg d.w. was comparable to the values found in city parks in Havana (87 mg/kg dw) and urban soils in general in Torino (90 mg/kg d.w.) (Rizo et al. 2011). Our result was higher than those in urban soils in general in Ljubljana, Belgrade, and Seville, but much lower than those in Thane in India and in Ankara in Turkey (Rizo et al. 2011). Our result was much higher than the level found in soils in Hualpen City, Chile (24 mg/kg d.w.) (Tume et al 2019). Regarding Zn (95.1 mg/kg d.w.), our result is much lower than that found in the same city parks in Havana (161 mg/kg d.w.) or other urban soils in general, as mentioned in Rizo et al. (2011), and is only comparable to the lowest values mentioned for Missouri and

Seville (95 and 105 mg/kg d.w, respectively). Our result was higher than that in soils in Hualpen City, Chile (59 mg/kg d.w.) (Tume et al 2019). Compared to similar studies performed in urban soil environments in Greece, our values were much lower (for Zn) or lower (for Cu) (Massas et al. 2009), similar (Golia et al. 2021), or slightly higher (Bourliva et al. 2021). It is worth mentioning that in 2010, playground topsoils in the much bigger city of Athens exhibited lower mean values of 43.4 mg/kg d.w. and 174.3 mg/kg d.w. for Cu and Zn, respectively (Massas et al. 2010).

The translation of these results into a risk to residents is shown in Fig. 3, which shows that there is acceptable risk to both adults (Fig. 3A) and children (Fig. 3B) through oral exposure in all cases. Nevertheless, the risk is more than 10 times higher for children than for adults. Furthermore, there are three "hotspots" with continuously elevated risk for Cu, while two sites showed the highest risk for Zn. All these sites are found towards the central-west part of the city, near the train station, where lower-income families and many immigrants tend to dwell, while higher-income families traditionally dwell closer to the Thessaloniki seafront. In general, green spaces in this city center are already scarce (Moussiopoulos et al. 2010) and are found mainly on its east side, besides the Seich-Sou grove at the city's outskirts, and are well below the acceptable standard of 9 m^2 of green space per city dweller (Latinopoulos 2022). Even though the main threat to both children and adults through the oral route remains the consumption of polluted grains and vegetables (Noubissié et al. 2016; Singh et al. 2018), soil eating, whether accidental or intentional, is extremely relevant for children (Sánchez-Nazario et al. 2003; Guney et al. 2010; Tume et al. 2019), who may play in these open spaces since they have nowhere else to go.

Regarding the dermal exposure, there is an acceptable risk for both adults and children, as shown in Fig. 4. Nevertheless, the risk is more than two times higher for children

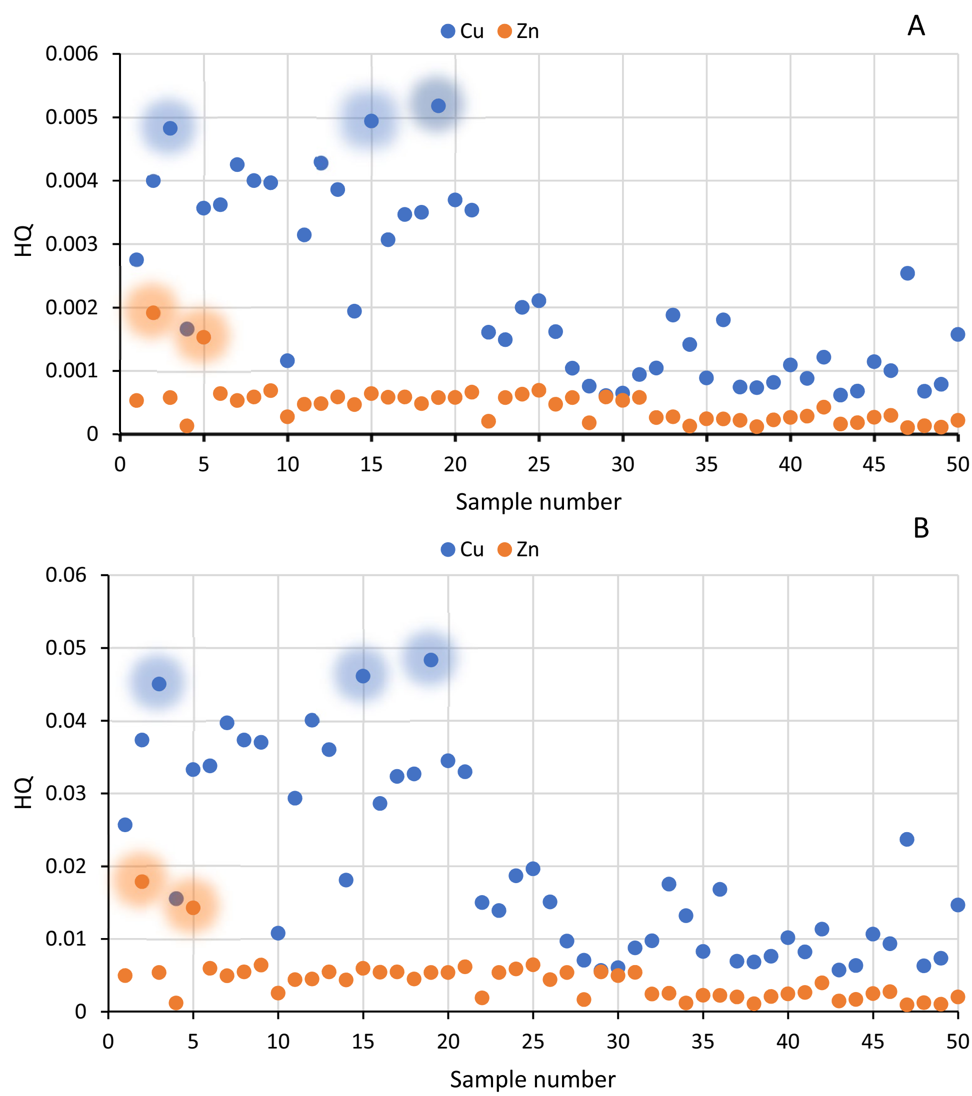


Fig. 3 Hazard quotient (HQ) values for the oral route of exposure of adults (**A**) and children (**B**) to soils from Thessaloniki. Each *blue dot* represents a risk for Cu and each *orange dot* represents a risk for Zn for each site ($n = 50$). Outliers for Cu and for Zn are drawn with a *halo*

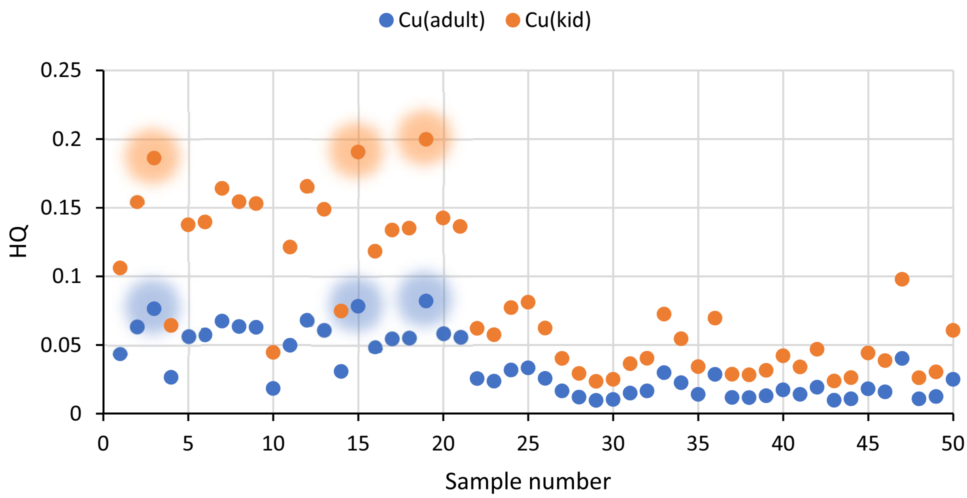


Fig. 4 Hazard quotient (HQ) values for the dermal route of exposure of adults and children to Cu in soils from Thessaloniki. Each *blue dot* represents a risk to adults and each *orange dot* a risk to children for each site ($n = 50$). Outliers for adults or children are drawn with a *halo*

than for adults. Furthermore, the three aforementioned hotspots show a continuously elevated risk for Cu. These samples were mainly from the area around the central train station and behind the station in the northwest direction. As discussed later, Cu and Zn were correlated; this means that soils polluted with Zn were also polluted with Cu, pointing to a possible source of contamination. It is known that in urban environments Cu mainly comes from wear to motor vehicle braking systems, especially from the frequent starting and stoping of road traffic (Li et al 2022), while Zn is a metal commonly found in the proximity of train stations (Golia et al. 2021). As mentioned in Wan et al. (2016), the railway station of Shijiazhuang City produced the most polluted dust, especially in Zn and Cu. According to the authors, this can be attributed to the heavy traffic to and from the station, the tearing of metal automobile and train parts, and friction with the overhead cables of trains, which can emit Cu. Furthermore, Thessaloniki is the largest commercial and passenger port of Northern Greece. The contribution of port activities to the metal pollution in the area is self-evident and has also been observed in other cities with busy ports (Aslanidis and Golia 2022). It is also important to note that other metals may also be present, so the real risk, which is additive, may be even higher than that calculated here.

The Spearman correlation results show a significant negative correlation between pH and EC ($R^2 = -0.722$, $p < 0.01$), which means that at a lower pH there is a higher EC (Nur Aini et al. 2014). There were higher concentrations of Cu and of Zn in the sandier soils ($R^2 = 0.673$, $p < 0.01$; $R^2 = 0.352$, $p < 0.05$ for Cu and Zn, respectively) and, conversely, lower concentrations in soils with more clay ($R^2 = -0.655$, $p < 0.05$; $R^2 = -0.401$, $p < 0.05$ for Cu and Zn, respectively), which is unusual since sandy soils have a low adsorption capacity (Tume et al. 2019). Clay shows a high sorption capacity and a strong ability to bind metallic elements (Kabata-Pendias 2011; Alloway 2013; Zaky and Abdel-Salam 2020). However, as mentioned by Zaky and Abdel-Salam (2020) and Giannakis et al. (2021), many other soil parameters may affect this capacity. Finally, Cu and Zn were highly positively correlated to each other, probably because of common sources ($R^2 = 0.605$, $p < 0.01$).

Conclusions

Urban soils may be enriched with several pollutants, posing health threats to the city dwellers. In this context, the present study showed the spatial distribution of Cu and Zn pollution in soil, where the Cu and Zn were of anthropogenic origin. The samples were taken from public green spaces found in the city center of Thessaloniki. Out of 50 samples, 1 sample was above the EU set limit for Cu and 2 samples were above the EU set limit for Zn in agricultural soil fertilized with sewage sludge according to 86/278/EEC. When compared to other guidelines for urban soil quality, a few samples were above the set limit for Cu and fewer samples were above the set limit for Zn. The risk for residents (adults and children) through the oral and dermal routes was acceptable in every case, but the risk for children, who are most likely to play in these green spaces, was ten times and two times higher than the risk for adults through the oral and dermal routes, respectively. The correlation between Cu and Zn points to common sources, which, given the locations of the sample areas, can probably be attributed to automobile congestion and train utilization. The possibility of concomitant pollution with other toxic metals in these areas should also be investigated.

Author contributions AC, AK: experiments, CE: writing—original draft, statistical analysis; EEG: conceptualization, supervision; AK: supervision. All authors have read and approved the final manuscript.

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Data availability The datasets analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of interest All authors declare that they have no conflict of interest.

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